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Short Communication

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Biomass burning emissions contaminate winter snowfalls in urban Beijing: A case study in 2012

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ABSTRACT

Three monosaccharide anhydrides levoglucosan, mannosan and galactosan were detected in winter snowfall samples of 2012 in urban Beijing. Concentrations of three isomers vary from 0.15 to 54.43 ng mL^{-1}, with an average value of 10.49 ng mL^{-1}. Levoglucosan is the most abundant component. Winter snowfalls are contaminated by biomass burning emissions seriously in urban Beijing. The main sources are softwood and crop residue burnings around Beijing from late autumn to early winter, while long–range transport of biomass burning emissions contribute more during the late winter. Concentrations of monosaccharide anhydrides in snowfall samples may be affected by both topography and the meteorological conditions around urban Beijing.

Keywords: *Biomass burning aerosols, levoglucosan, snow chemistry, air pollution, urban Beijing*

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1. Introduction

Air pollution has been a widespread topic in recent years (Akimoto, 2003; Huang et al., 2014). Aerosols and greenhouse gases are the two main air pollutants (Akimoto, 2003). Aerosols can significantly alter the Earth's radiation by scattering and absorbing solar radiation, and impact microphysical characteristics of cloud droplets (Ramanathan et al., 2001; Gyawali et al., 2009). Atmospheric Brown Clouds (ABCs) caused by anthropogenic aerosols in South Asia has been received public attention due to the unclear effects on regional and global climate system and hydrological cycle (Ramanathan et al., 2001; Ramanathan and Crutzen, 2003). Similar phenomena were also observed at densely–populated regions like Central America, Brazil, the Mediterranean and North China in recent years (Ramanathan and Crutzen, 2003; Li et al., 2010).

Biomass burning (BB) emissions contribute to more than half of the carbonaceous aerosols (including both elemental carbon and organic carbon) all over the world (Jacobson et al., 2000; Gyawali et al., 2009). This is more serious in China, e.g. urban Beijing (Li et al., 2010; Cheng et al., 2013). Several studies suggested that air quality in Beijing was impacted significantly by BB emissions all year around, and winter was the most remarkable time (Wang et al., 2006a; Cheng et al., 2013; Yu et al., 2013). Burning of crop residue (include wheat–straw and cornstalk) and firewood were highlighted as two main sources of organic aerosols in urban Beijing from late autumn to winter (Wang et al., 2009; Yu et al., 2013). Aerosol samples collected from urban Beijing showed high concentrations of organic matter during winter time (Wang et al., 2006a; Zhang et al., 2008; Huang et al., 2014). The organic matter in aerosol particles has a complex composition in urban Beijing. More than one hundred kinds of organic compounds were identified in aerosol samples collected from urban Beijing (Wang et al., 2006a; Wang et al., 2006b). Polycyclic aromatic hydrocarbons (PAHs), n–alkanes, fatty acids and alcohols, polyols/poly acids, dicarboxylic acids and saccharides have been considered to be the main organic components (Wang et al., 2006b). Among various kinds of organic compounds, levoglucosan and its isomers showed similar variation patterns to traditional BB tracer K^+ all year around (Cheng et al., 2013). Levoglucosan accounts about 90% of total identified sugars in winter aerosols in urban Beijing (Zhang et al., 2008; Cheng et al., 2013). These studies confirmed that levoglucosan and its isomers could be useful biomarkers for BB studies in urban Beijing.

However, previous studies mainly focused on records of monosaccharide anhydrides (MAs) in aerosols (Wang et al., 2006b; Zhang et al., 2008; Cheng et al., 2013). Considering of their specific sources and characteristics, these MAs can be ideal biomarkers for BB studies in aqueous samples (You et al., 2014). In this study, levoglucosan, mannosan, galactosan were detected simultaneously using Gas Chromatography/Mass Spectrometry (GC/MS) method in winter snowfall samples collected from urban Beijing. The possible impact of BB on snowfall samples in urban Beijing was also analyzed.

2. Experiment and Methods

Snow samples were collected at The Campus of Institute of Psychology, Chinese Academy of Science (40°00'N, 116°22'E). The sampling site is located at central lawn of the campus, and there are no obvious BB sources within 5 km around. Fresh snow was collected in a plastic container with polyethylene (PE) bags when it began to snow. Samples were compacted into pre–cleaned polyethylene terephthalate (PET) bottles within half an hour after snowfall stopped. There were four snow samples collected in total from November 2012 to January 2013 (please see detailed sample information in Table 1). Samples were kept frozen at a temperature of –20 °C before analysis. Samples were dried by freeze–drying for at least 24 h, and then extracted three times using an ultrasonic method for 30 min in a 5 mL mixture of dichloromethane/ methanol=4/1 (v/v). After concentrated and dried completely under high purity N_2, samples were silylated by N–methyl–N–trimethylsi–lyltrifluoroacetamide containing 1% trimethylchlorosilane before GC/MS analysis. Silylated extracts were analyzed using a Trace GC 2000 gas chromatograph connected to a PolarisQ ion trap mass spectrometer. More detailed information about the sample preparation and determination can been found in a previous study (You et al., 2014).

Table 1. *Meteorological data in four snowfall events*

	T [a] (°C)	RH [b] (%)	SEW [c] (mm)	WS [d] (m s^{-1})	WD [e]
Nov 4th 2012	2.9	82	48.9	5.2	N [f]
Dec 12th 2012	–2.1	69	0.2	1.6	SWW [g]
Dec 21st 2012	–4.6	71	0.6	0.8	SWW [g]
Jan 20th 2012	–2.5	72	2	0.9	SE [h]

[a] *Temperature,* [b] *Relative humidity,* [c] *Snow equivalent water,* [d] *Wind speed,* [e] *Wind direction,* [f] *North,* [g] *South West West,* [h] *Southeast*

Meteorological data is from the China Meteorological Administration. To identify the BB source regions more clearly, backward trajectories were calculated by HYSPLIT model and MODIS hotspot data (1 km×1 km, AFD, 2013) were used (Justice et al., 2002). Because the lifetime of BB aerosols are usually no more than a week (Ramanathan and Crutzen, 2003), seven days backward air mass trajectories were used in this study. The matrixes (centered with sample site, 0.1°×0.1°, 9 points) instead of single–points were used to identify the potential about BB sources around sampling site in HYSPLIT model. The height of the atmospheric boundary layer (ABL) varied from 300 to 600 m above ground level (AGL) in urban Beijing during winter time (Guinot et al., 2007). The height of 100 m (green line), 500 m (blue line) and 3 000 m (red line) AGL was set to analyze the backward trajectories in ABL, the top of ABL and above ABL, respectively.

3. Results and Discussion

3.1. BB aerosols contaminate winter snowfall samples

Concentrations of three isomers in winter snowfall samples ranged from 0.15 to 54.43 ng mL^{-1}, with an average value of 10.49 ng mL^{-1}. Levoglucosan with an average value of 25.30 ng mL^{-1}, accounts for more than 80% of the total detected MAs. Detailed results can be seen in Table 2. Levoglucosan, mannosan and galactosan can only be generated from the degradation of cellulose and hemicellulose when the burning temperature is higher than 300 °C (Simoneit et al., 1999). Therefore, those three MAs are specific biomarkers for BB aerosols. The existence of those three MAs indicates that winter snowfall samples in urban Beijing are substantially contaminated by BB emissions.

Previous observation and modeling results indicated that wet deposition was the dominant sink for organic matter, especially at middle and low latitude regions (Hu et al., 2013). The following equation is used to transform the concentrations of MAs in snow samples to their related concentrations in the atmosphere (Davidson et al., 1993):

$$C_a = \rho_a \times C_s/\omega \tag{1}$$

where C_a is concentration of species (ng m^{-3}) in the atmosphere, ρ_a is air density (g m^{-3}) under the standard temperature and pressure (0 °C and 1.01×10^5 Pa, in this study the value of 1 293 g m^{-3} is used), C_s is concentration of MAs (ng g^{-1}, actually equals to ng mL^{-1}) in snow, and ω is the scavenging ratio for specific species by snowfall. The value 125 was recommended as a worldwide mean of ω for both black carbon and organic matter (Jacobson, 2004), and this value is used to estimate the concentrations of MAs approximately in this study.

Concentrations of MAs in the atmosphere estimated by the Equation (1) in four snowfall events are shown in Table 2. Total concentrations of MAs varied from 16.01 to 683.15 ng m^{-3}, with an average of 325.61 ng m^{-3}. Levoglucosan concentrations ranged from 6.86 to 563.07 ng m^{-3}, with an average of 261.65 ng m^{-3}. Results in this study can be comparable to previous aerosol studies in urban Beijing. Concentration of levoglucosan was about 575 ng m^{-3} in $PM_{2.5}$ aerosol samples in winter (Zhang et al., 2008). Concentrations of levoglucosan varied from 60 to 1 940 ng m^{-3}, with a mean value of 590 ng m^{-3}, and concentrations of mannosan varied from 50 to 190 ng m^{-3} in typical winter aerosol samples (Cheng et al., 2013). However, the estimated concentrations of MAs in this study are much higher than previous aerosol studies at other regions in China. Concentrations of levoglucosan in aerosols at Mount Tai during the wheat straw burning period varied from 88 to 1 210 ng m^{-3}, with an average of 403 ng m^{-3} (Fu et al., 2008). Concentrations of levoglucosan in winter aerosols in Guangzhou varied from 57.9 to 269.3 ng m^{-3}, with an average value of 176.6 ng m^{-3} (Ma et al., 2009). The concentrations of levoglucosan at Hong Kong ranged from 26.2 to 133.7 ng m^{-3} (Sang et al., 2011). Those comparable results evidently proved that snowfall samples in urban Beijing were seriously contaminated by BB emissions.

3.2. Identification of BB sources

Previous studies suggested different ratios of MAs (e.g. levoglucosan/mannosan) can be useful to identify the BB sources in urban Beijing (Cheng et al., 2013). Although degradation happens during the transport process, the impact of the degradation on the ratios among MAs has been considered to be limited (Hu et al., 2013). Different snow samples display distinct ratios of levglucosan/ mannosan, indicating that snowfall samples in urban Beijing are contaminated by various sources of BB in winter (Table 2 and Figure 1). The ratio of levoglucosan/mannosan is 10.32 for December 12th and 8.86 for December 21st 2012, respectively. The ratios are consistent with results of aerosol studies in Beijing (levoglucosan/mannosan with an average of 9.01±1.47 for typical winter samples) (Cheng et al., 2013). Snowfall samples on December 12th and December 21st 2012 may be contaminated by mixed aerosols released from softwood and crop residual (Figure 1). Hardwood, grass and leaf burning may also act as non–negligible sources. However, ratios of levoglucosan/mannosan on November 4th 2012 and January 20th 2013 are different from previous aerosol studies, which indicate snowfalls on these two days are contaminated by different BB emission sources. Compared with previous studies (please see detailed data in the Supporting Material, SM), the possible sources are soft wood, needle and duff burning on November 4th 2012 and snowfall on January 20th 2013 may be contaminated by hardwood burning. This classification method may have some uncertainties, because it is based on limited statistical analysis results from previous studies. However, this method can help us to understand the contaminations of snowfall samples caused by BB aerosols at some extent.

Table 2. *Concentrations of MAs in four snowfall samples (ng mL^{-1})*

	Nov 4th 2012	Dec 12th 2012	Dec 21st 2012	Jan 20th 2013
Galactosan	0.73	4.99	5.47	1.66
Mannosan	0.15	4.39	6.14	1.20
Levoglucosan	0.66	45.31	54.43	0.77
Levogluocsan/Mannosan	4.41	10.32	8.86	0.64
Estimated concentrations of MAs in the atmosphere for four snowfall events (ng m^{-3})				
Galactosan	7.60	51.64	56.54	17.17
Mannosan	1.56	45.41	63.54	12.41
Levoglucosan	6.86	468.71	563.07	7.95
Total MAs	16.02	565.76	683.15	37.53

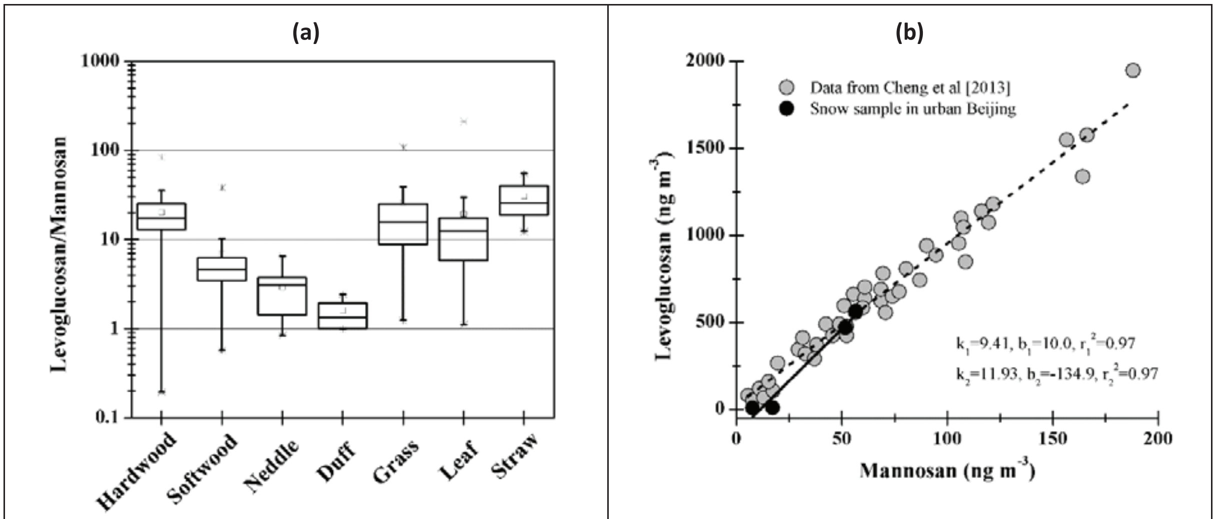

Figure 1. (a) *Levoglucosan/mannosan ratios among various kinds of BB sources from different references. The central bar is the median, and the box shows the first and the third quartiles, respectively. The whisker shows the maximum and minimum values. The hollow block symbols indicate arithmetical means. The crosses indicate extreme values. Detailed data can be found in the SM.* ***(b)*** *The dependence of levoglucosan on mannosan in winter samples in urban Beijing, the gray dots (k_1, b_1, r_1^2) are aerosol samples from Cheng et al. (2013), the black dots (k_2, b_2, r_2^2) are snow samples in this study.*

The results based on HYSPLIT backward air mass trajectories indicate potential emission source regions (Figure 2). The northeastern China is seen as the major source region for BB aerosols near the surface (100 and 500 m) on November 4th 2012, while the Jiaodong Peninsula and the Eastern Coastal China are seen as the two major source regions at high altitude (3 000 m, Figure 2a). During late September to early November, the atmosphere is greatly influenced by the burning of agriculture crop residue and fallen leaves in Northern China. Burning fumes can be transported above the ABL, and thus can be transported to regions far away from the emission sources. Similar records of crop residue burning were reported by previous studies in urban Beijing and regions nearby (Zhang et al., 2008; Cheng et al., 2013). The dry weather is prevalent since late autumn at most regions of northern China (Zhai et al., 2005), which increases the risks of forest and grassland fires. The BB source regions for two snowfall samples in December 2012 are different from snowfall on November 4th 2012 (Figure 2b and 2c). BB aerosols mainly release from Northern China and Mongolia and sometimes from as far as central Asia in December 2012. Although only few forest or steppe fires occur in winter time at those regions, firewood burning for heating and cooking are prevalent in northern China, Mongolia and central Asia in winter (Wang et al., 2006a; Zhang et al., 2008). It is also noticeable that BB emissions from Northeastern China may be one of the most important sources for snowfall on December 12th 2012, especially for aerosols transported at low altitude (100 and 500 m AGL). The result based on HYSPLIT backward air mass trajectories indicates that BB contaminations on January 20th 2013 are from distinct regions at different altitudes (Figure 2d). Pollutants mainly come from northern China and nearby regions at the low layers (100 and 500 m), while pollutants may come from southwestern China at 3 000 m AGL. Southwestern China is mainly covered by large areas of broad–leaved deciduous forest and bunchgrass. Fires occur during January 2013 at Yunnan, Sichuan, Henan and some other provinces (XHN, 2013) along the trajectories due to the dry weather conditions. Additionally, burning incense and joss paper during the Spring Festival is prevalent in those regions, which can also contribute to long–range transport of BB aerosols.

3.3. Possible impact factors

Previous studies suggested that aerosols released from BB could persist in the atmosphere from hours to several days (China et al., 2013). The transport pathways are important to understand the effects of BB aerosols on snowfall samples in urban Beijing. Urban Beijing is located at the foothill of northeast–southwest oriented Yanshan Mountains. Prevailing winds are mostly from southeast and southwest during the investigated period (Table 1). This is consistent with result of HYSPLIT model. The orientation of mountain provides possible pathways for BB aerosols. The distribution of surrounding mountains form the semi–basin topography of urban Beijing, which facilitates the accumulation of BB aerosols (Wu et al., 2011). The accumulated BB aerosols can lead to higher concentrations of MAs in urban Beijing.

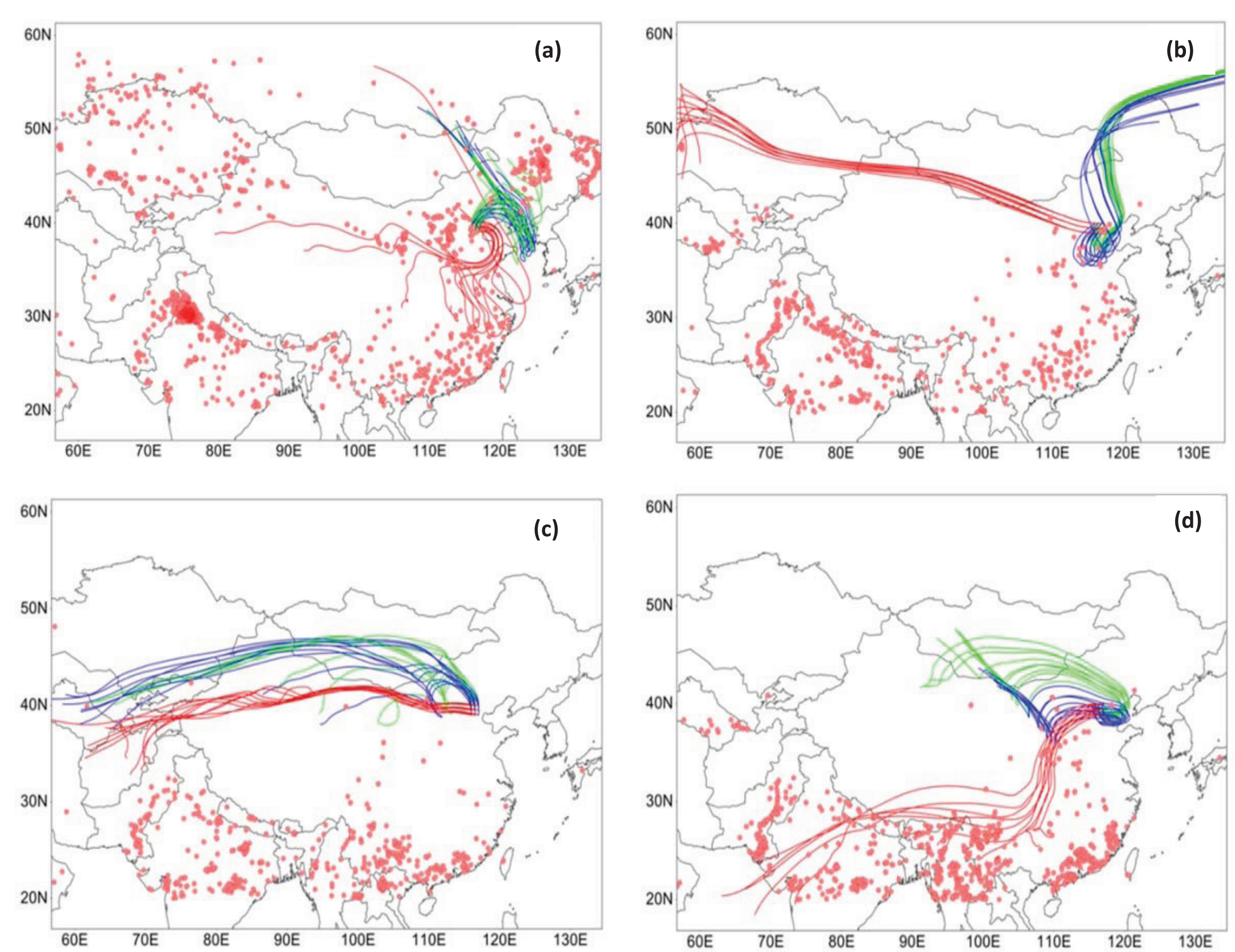

Figure 2. *Seven-day backward air mass trajectories based on the HYSPLIT model for BB sources identification. The green lines stand for 100 m AGL, blue lines stand for 500 m AGL and red lines stand for 3 000 m AGL. The red dots are fire hotspots from MODIS.* ***(a)*** *November 4th 2012,* ***(b)*** *December 12th 2012,* ***(c)*** *December 21st 2012 and* ***(d)*** *January 20th 2013.*

The dispersion of BB aerosols is influenced by many other factors in addition to emission sources. The meteorological conditions are thought to be the most important factors which affect the diffusion process (Hu et al., 2013; Sun et al., 2013). Due to limited samples, we can only give the extrapolation but not statistical results. High concentrations of MAs in snow samples occur with low precipitation and low wind speed (Figure 3). This might be because snowfall can dilute the BB aerosols, which leads to lower concentrations of MAs in snow samples. Previous studies of marine aerosols found that concentration of levoglucosan was strongly influenced by the effect of precipitation scavenging at low latitude regions (Hu et al., 2013). The dispersion of BB aerosols is faster in windy days than calm days (Sun et al., 2013), leading to higher concentrations of MAs in calm days. Previous studies indicated that faster decay of levoglucosan was observed at higher relative humidity conditions in laboratory (Hennigan et al., 2010). Concentrations of MAs in snow samples and relative humidity show an inverse variation trendency in urban Beijing during the study period (Figure 3). This result indicates that relative humidity might even affect the oxidation of MAs after deposition, which is important to understand the effectiveness of levoglucosan as biomarker in sediments. However, this hypothesis is deduced by preliminary results in this study, which needs more data to prove it.

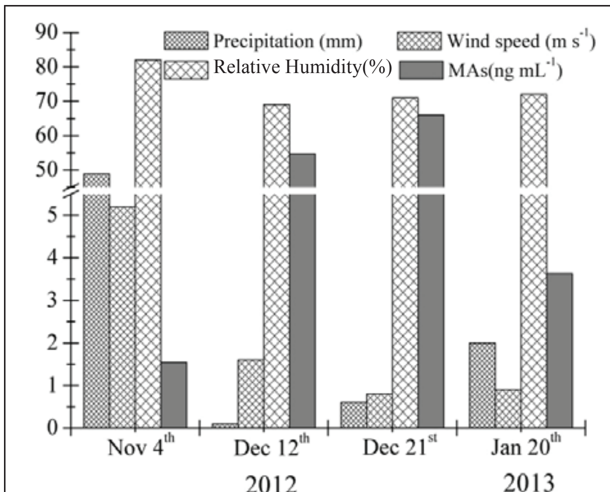

Figure 3. *Influence of major meteorological factors (precipitation, wind speed and relative humidity) on concentrations of MAs in four snowfall events.*

4. Conclusions

This study reported BB signals in snowfall samples using MAs records in urban Beijing during 2012 winter period. Snowfall samples are contaminated by both regional emissions and long–range transport aerosols released from various kinds of BB. The possible BB sources are revealed by ratios of levoglucosan/ mannosan, HYSPLIT model and MODIS fire hotspots. The concentrations of MAs show significant temporal variations. The topography may affect the pathways of BB aerosols. Meteorological conditions may affect the dispersion of BB aerosols and the concentrations of MAs in snowfall samples. This study is helpful to understand the contribution of BB emissions to air pollution in urban Beijing, and is also helpful to the environmental significance of BB emissions.

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Supporting Material Available

Ratios among three monosaccharide anhydrides (MAs) from different sources (Table S1). This information is available free of charge via the internet at http://www.atmospolres.com.

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